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(54) Deep-drawable films, methods for their manufacture, and use of same

(57) The present invention relates to a deep-drawable multilayer film that contains a top film with certain parts by weight of a) at least one crosslinked PP-EPDM and b) an uncured propylene block copolymer or heterophase propylene block polymer, wherein 1 to 15 wt% of b) is substituted by the same parts by weight of a polymer and/or ionomer c) containing a reactive group.

The bottom film or films contains/contain at least two different polyolefins or olefin-group-containing compounds.

The following specifications have been taken from the documents submitted by the applicant

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## Specification

The present invention relates to a deep-drawable multilayer film that contains a polyolefin-containing top film and at least one polyolefin-containing bottom film that deviates qualitatively and/or quantitatively from the composition of the top film, as well as, optionally, a surface protection layer wherein the top film [contains] certain parts by weight of a) a crosslinked PP-EPDM and b) an uncured propylene block copolymer or heterophase propylene block polymer, wherein b) is partially substituted by a polymer and/or ionomer c) that contains a reactive group.

The bottom film contains at least two different polyolefins or polyolefin-containing polymers, for example, partially crosslinked EPDM, polypropylene and/or reactive-group-containing polymers and/or ionomers and/or polymeric crosslinking agents or one or more of the polyolefin-containing polymers named above and an ethylene-vinyl acetate copolymer.

A tremendous variety of polyolefin-based films, particularly those based on polypropylene along with polyethylene and its processed forms are already known (see FR-A-2017705, among others). However, the demands on the films, depending on the area of application, are so different and are associated with increasing demands, so that there is a demand for films with improved *[missing word in original document]* and/or improved processing possibilities.

In the context of this invention, numerous experiments with film formulations were carried out in which, among other things, such formulations were tested, containing crosslinked PP-EPDM or an uncured propylene block copolymer or heterophase propylene block polymer.

These films exhibit a certain scar\* resistance and have a degree of dullness that is still sufficient in several areas of application. Other possibilities for providing plastic film surfaces with a matte finish with matte paints are not suitable for the production of deep-drawn films or do not yield any uniform structures with matte effects after embossing.

Special demands are to be adhered to for the production of deep-drawable automobile films, sheeting, or automobile film layers, since different deep-drawing conditions have to be used with the same molded part. Nevertheless, the molded part is supposed to exhibit the same embossing structure and matte finish throughout. On the other hand, special demands are to be adhered to regarding the scratch resistance of the plastic surfaces, fog values, aging stability, and the like.

It was the goal or objective of the present invention to achieve, in comparison with the prior art, an improved film with improved *[deep-drawing]* characteristics or processing

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\* [Literal translation; later in the document, the German word for "scar" is stated as something that is desirable.]

characteristics. In particular, the film was to exhibit a good or improved scar resistance, and the surfaces were to be matte or exhibit a certain dullness or degree of dullness. In particular, however, the film was to have good deep-drawing characteristics and also be able to be processed into molded parts or objects onto which relatively high demands are placed.

Such areas of application are, for example, molded parts, objects, or deep-drawn parts for automobiles, particularly interior automobile trim.

According to the invention, it was determined that a deep-drawable multilayer film satisfies these goals or objectives, which involves the use of a polyolefin-containing top film and at least one polyolefin-containing bottom film that differs qualitatively and/or quantitatively from the composition of the top film, as well as, optionally, a top surface protection layer.

According to the invention, the top film contains (with respect to 100 parts by weight of surface film plastic)

a) 20 to 80 parts by weight

of at least one crosslinked PP-EPDM (alloy or mixture of a propylene homo- or copolymer and crosslinked or partially crosslinked ethylene-propylene diene copolymer)

b) 20 to 80 parts by weight

of an uncured propylene block copolymer or heterophase propylene block polymer with one elastomer component in the block or in the chain of [a] –5 to 35 wt% (with respect to 100 parts by weight of the uncured heterophase propylene block copolymer or heterophase propylene block polymer), wherein 1 to 15 wt%, preferably 2 to 10 wt%, of b is substituted by the same parts by weight of a polymer and/or ionomer c that contains a reactive group, wherein

c) at least one polymer and/or ionomer c that contains a reactive group is based on ethylene and methacrylic acid and/or acrylic acid, wherein the acid groups partially contain metal ions or are neutralized by metal ions.

The bottom film or films contains/contain at least two different polyolefins or polyolefin- or olefin-containing polymers, co-, ter-, graft, or block polymers.

The multilayer film according to the invention is a flexible, deep-drawable automobile film or sheeting that is particularly well suited for the manufacture of thermoformed molded parts with uniformly matte surfaces for automobile films used for interior trim.

The dullness [matte surface] of this film, to a large extent, is independent of the deformation temperature and the degree of deformation, that is, there is uniform or nearly uniform dullness of the film surface throughout the molded part or the thermoformed molded part made from it. The top film here preferably contains a dynamically crosslinked or partially crosslinked PP-EPDM or PP-EPDM mixtures or alloys. Components a), b), and c) of the surface are optionally used together with processing aids, adjuvants, fillers, coloring agents, and/or color pigments.

The polypropylene block of the uncured heterophase polypropylene block polymer preferably consists of a propylene, co-, and/or block polymer. The elastomer component of the heterophase polypropylene block copolymer, according to a preferred embodiment, is 40 to 70 wt% (with respect to 100 parts by weight of the uncured heterophase polypropylene block copolymer). However, according to another preferred embodiment, the elastomer component of the uncured heterophase polypropylene block copolymer is present at 35.5 to 48 wt%, preferably 36 to 45 wt% (with respect to 100 parts by weight of the uncured heterophase polypropylene block copolymer). According to a preferred embodiment, the elastomer of the ethylene propylene copolymer contains an ethylene component at 20 to 70 wt%, preferably 30 to 60 wt%, and a propylene component of 30 to 80 wt%, preferably 40 to 70 wt%, and has a molecular weight greater than 100,000, preferably greater than 150,000.

The addition of a dynamically crosslinked PP-EPDM blend in the top and/or the bottom film, with the particle size of the crosslinked elastomer phase being 0.5 to 15  $\mu\text{m}$ , improves the deformation range [elasticity], the uniformity of the thermal expansion, and the scar characteristics. Improved scar characteristics mean that the over-proportional expansion of the scar valleys [depth of the scars] throughout the solid object, which increases as the expansion increases, is limited (strain hardening). In order to achieve this effect, a homogeneous working-in of the crosslinked PP-EPDM blend and a particle-form elastomer structure are necessary.

Certain demands with respect to the degree of crosslinking and the melt index of the crosslinked PP-EPDM blend used arise as a result of this. The degree of crosslinking in complex systems, indirectly defined through the compression set at 70°C after 22 h (DIN 53517), should lie between 25 and 60.

A block polymer with an Mfi 230/2.16 g/10 min of 0.3 to 8, preferably 0.5 to 7, is preferably used as the heterophase propylene block polymer.

Dynamically crosslinked or partially crosslinked PP-EPDM, preferably those having a density between 0.83 and 0.96 g/cm<sup>3</sup>, preferably 0.87 and 0.91 g/cm<sup>3</sup>, are preferably used for the top and/or bottom films as the PP-EPDM. Preferably, the PP-EPDM selected for this has a flexural modulus (initial flexural modulus) – measured according to ASTM D 790 with a 2-mm-thick film sheet – of 80 to 400 MPa, preferably 100 to 300 MPa.

The deep-drawable multilayer films according to the invention possess a degree of gloss, according to DIN 57530/600, of 0.5 to 5, which is essentially influenced neither by the required deformation temperature nor by the deep-drawing conditions. This characteristic thus distinguishes the films according to the invention with regard to the knowledge about films collected in the past. Also, these films are matted through the use of fillers or embossing.

Using the additive according to the invention or the mixture according to the invention for the manufacture of deep-drawable automobile films, it is possible to obtain plastic films with a matte surface and high elasticity after thermodeformation. In so doing, the thermal molding of the multilayer film according to the invention can be carried out over a broad temperature interval of approximately 140 to 200°C, preferably 150 to 185°C, without the occurrence of particular difficulties.

The film has an outstanding deep-drawability and can be used both in positive deep-drawing methods and negative deep-drawing methods. Using this composition, it is possible to obtain deep-drawing films with a Shore D hardness of 25 to 60, preferably 30 to 55, and a degree of gloss according to Gardner 60°C [sic] of 0.5 to 1.5, measured on the deep-drawn molded part.

Moreover, the multilayer automobile films for interior trim according to the invention possess the advantage of a high aging stability under light and heat, as well as low fog values.

According to a preferred embodiment, the top and/or bottom film has a filler content of 0 to 30 parts by weight, [preferably] 0 to 20 parts by weight (with respect to 100 parts by weight of plastic). As fillers, those that are in themselves known, preferably talc and/or SiO<sub>2</sub> with an average particle size of 0.01 to 20 µm, preferably 0.05 to 10 µm, can be used.

Among other things, heat and/or light stabilizers for polyolefins are preferably used as processing adjuvants.

Among other things, lubricants or lubricant mixtures are preferably used as other processing adjuvants.

According to one embodiment, an adhesion promoter layer is preferably arranged underneath the film layer. The adhesion promoter layer preferably consists of acrylates, polyurethane or acrylate polyurethane mixtures, or mixtures containing them. A foam layer, a substrate layer, a tissue layer, and/or supporting layer is preferably arranged underneath the film layer or adhesion promoter layer. The foam layer preferably contains a polyolefin foam or a polyurethane foam, or a mixture containing such.

Preferably, the top film has an average thickness of 60 to 500 µm, preferably 100 to 350 µm. The bottom film or films has/have an average thickness of 100 to 2000 µm, preferably 500 to 1500 µm; also, the bottom film or films has/have a total thickness that is more than 1.5 times, preferably 2 or more times, greater than that of the top film.

The multilayer film according to the invention has a very good thermo-formability and is used for the manufacture of low-tension molded parts with a precise rendering or reproduction, or with form- or temperature-stable surface structures. The multilayer film is preferably processed in positive deep-drawing methods and/or negative deep-drawing methods. On its underside or bottom surface, the multilayer material preferably has a foam layer, for example, polyurethane foam, but preferably polyolefin foam, and/or it is provided with a substrate layer or a substrate and/or a supporting layer and/or a lattice or tissue layer. According to a preferred embodiment, the film or layer [sic] is provided with decorative features, scars, embossings and the like, is scar-resistant, easily gripped, and can be printed on. Using the multilayer film according to the invention, it is possible to produce molded parts or objects whose surfaces exhibit a good scar stability even in thermal storage.

According to a preferred embodiment of the invention, the bottom film contains at least one partially crosslinked EPDM and a propylene homo-, copolymer or graft-polymer with or without reactive groups, a PP-EPDM or dynamically crosslinked or partially crosslinked PP-EPDM with at least one of the components b or c of the top film and/or at least one polymer and/or ionomer containing a reactive group, or a mixture thereof.

According to another preferred embodiment, the bottom film consists (with respect to the plastic component) of 65 to 90 wt% (with respect to 100 wt% plastic of the bottom film) a propylene homo-, co-, ter-, and/or block polymer and 10 to 35 wt% ethylene-vinyl acetate copolymer with a vinyl acetate content of 10 to 40 wt%, preferably 20 to 35 wt%, wherein a plastic component or the mixture of the plastic alloy is treated with 0.1 to 5 wt%, preferably 0.2 to 1.5 wt% of at least one peroxide or a peroxide-group-containing compound. In so doing, the bottom film preferably consists of 70 to 85 wt%, preferably 75 to 83 wt% (with respect to 100 wt% plastic of the bottom film) propylene homo-, co-, ter-, and/or block polymer and 15 to 20 wt%, preferably 17 to 25 wt% ethylene-vinyl acetate copolymer.

According to another embodiment, the bottom film contains at least one partially crosslinked EPDM, a propylene homo-, copolymer, or graft polymer with or without reactive groups, as well as a block, di-block, or tri-block and/or graft polymer containing an additional olefin or polyolefin and/or styrene or polystyrene, or which is completely or partially substituted by a modification agent based on a polypropylene-maleic acid anhydride mixture or graft polymer, a polypropylene-acrylic acid mixture or graft polymer, an ethylene-vinyl acetate copolymer, an ethylene-acrylic acid or methacrylic acid copolymer, an ethylene-acrylic acid copolymer, an ethylene-acrylic acid ester copolymer, a glycidyl methacrylate co- or terpolymer, preferably an olefinic or olefin group-containing glycidyl methacrylate co- or terpolymer and/or ethyleneglycidyl methacrylate terpolymer or copolymer, an olefinic ionomer, a polyurethane, a polycaprolactone, a polyester ether, and/or an polyether amide.

Preferably, the components of the bottom film are selected in such a manner that the bottom film made thereof (with respect to the initial cross-section of the bottom film) has a maximum tensile stress in the range of  $20 \times 10^3$  to  $70 \times 10^3$  Pa and/or that the strain-stress curve runs approximately parallel to the elongation axis at an elongation of over 150%, preferably over 300% elongation after reaching the maximum [strain-stress].

The objects, molded bodies, parts, and the like manufactured or deep-drawn from this [film], which preferably contain a foam layer, substrate layer or a substrate [sic], supporting layer and/or a lattice or tissue layer, are preferably used for control panels or trim boards for automobiles, side wall parts for automobiles, door trim for automobiles, interior ceiling trim (inside roof lining), interior rear-wall parts for automobiles (rear shelf [compartment]), and other interior trim parts or objects for automobiles. According to a preferred embodiment, the substrate layer and/or foam layer contains polyolefins or is polyolefin-based.

The polymers and/or ionomers of the bottom film, which contain reactive groups, according to one embodiment, preferably contain alkali metal and/or alkaline earth ions and/or zinc ions as the metal ion.

According to one embodiment, the polymers and/or ionomers containing reactive groups contain alkyl acrylates and/or acryl methacrylates with 1 to 8 C atoms.

According to a preferred embodiment, the propylene or polypropylene content (content of propylene homo-, co-, ter- or block polymer) of the crosslinked PP-EPDM (component a) of the surface with respect to 100 parts by weight of the PP-EPDM contained in the top film is between 60 and 90 wt%, preferably 65 to 85 wt%, whereas the EPDM that has been crosslinked with a peroxide or a peroxide-group-containing compound possesses a part by weight of 10 to 40 wt%, preferably 15 to 35 wt%.

According to a preferred embodiment of the invention, the top film has a [precise] Shore D hardness or is adjusted to it [such a level], with this Shore D hardness being more than 3 Shore D units, preferably 5 to 10 Shore D units lower than the Shore D hardness of the bottom film(s) and/or with the surface tension of the top film being set to a value of greater than 40 mN/m.

Preferably, the top film using at least one plastic with a lower Shore D hardness and at least one component or plastic with a higher Shore D hardness is set to a Shore D hardness of less than 37, preferably to a Shore D hardness of 28 to 35, and the Shore D hardness of the bottom film(s) is at least 38 Shore D, preferably 39 to 48 Shore D.

According to another preferred embodiment, at least one polyacrylate, polymethacrylate, and/or polyurethane-containing layer is arranged above the top film or on the film surface and/or underneath the bottom film whose layer thickness is less than the layer thickness of the top film or bottom film, wherein the layer thickness of the coating is less than 50% of the layer thickness of the top or bottom film(s).

Here, the top film and, optionally, partial areas of the bottom film(s) are preferably embossed or scarred and provided with at least one polyacrylate, polymethacrylate, and/or polyurethane-containing layer, or a coating that contains 0.01 to 18 wt%, preferably 0.1 to 12 wt%, of at least one fine matting material and is coated by the application of a diluting agent.

The polyacrylate, polymethacrylate, and/or polyurethane-containing layer(s) or coating(s) or layer(s) or coating(s) consisting of them [sic] (with respect to the plastic component) preferably contain at least one antistatic agent and/or at least one adhesion-promoting chemical agent, preferably ultrafine carbon or soot with an average particle size of under 100 nm, preferably between 10 and 80 nm, and/or at least one ultrafine silicic acid or an ultrafine silicon dioxide and/or ultrafine silicate with an average grain diameter of under 4  $\mu\text{m}$ , preferably under 3  $\mu\text{m}$ .

According to another embodiment, the bottom film or films consist of (with respect to 100 parts by weight of the plastic of the bottom film or films):

1 to 60 wt%, preferably

5 to 55 wt%,

of at least one partially crosslinked EPDM

12 to 30 wt%, preferably

13 to 25 wt%,

of a propylene homo-, copolymer or graft polymer with or without reactive groups,

9.5 to 77 wt%, preferably

19.0 to 73 wt%,

of at least one ethylene-, methacrylic acid-, or acrylic acid-based polymer and/or ionomer that contains reactive groups, wherein the acid groups partially contain metal ions or are neutralized by metal ions, and

0.5 to 10 wt%, preferably

1.0 to 9 wt%,

of at least one polymeric crosslinking agent that contains reactive groups and contains more than 51 wt% (with respect to 100 parts by weight of the polymeric crosslinking agent) of ethylene or other olefin groups and from 1 to 49 wt% of acrylate and/or methacrylate groups and reactive groups, wherein epoxy, isocyanate, ketone, aldehyde, silane, alkyl halide, and/or anhydride groups are considered to be [desired] reactive groups.

According to a preferred embodiment, the polymeric crosslinking agent of the bottom film or films that contains reactive groups has, besides [*words missing in original document*] acrylate and/or methacrylate groups, or acrylic acid and/or methacrylic acid groups, and more than 51 wt% ethylene and/or olefin or ( $\text{CH}_2$ ) or CH groups, epoxide groups, or epoxy-like groups, preferably glycidyl groups. The polymers and/or ionomers of the bottom film, which



contain reactive groups, preferably contain, as a metal ion, alkali metal and/or alkaline earth ions and/or zinc ions. According to one embodiment, the polymers and/or ionomers that contain reactive groups contain alkyl acrylates and/or alkyl methacrylates with 1 to 8 carbon atoms.

According to another embodiment of the invention, the plastic film or plastic film sheet contains at least one bottom film and contains or consists of, as a plastic component or plastic mixture, (x) 5 to 80 parts by weight, preferably 10 to 60 parts by weight, of at least one polymer that contains elastic reactive groups or at least one elastomer that contains thermoplastic reactive groups or a polymer or polymer mixture that contains reactive groups containing a component of elastic polymers of more than 60 wt%, preferably more than 75 wt% (with respect to 100 parts by weight of the elastomer-containing polymer or polymer mixture), (y) 20 to 95 parts by weight, preferably 40 to 90 parts by weight, of at least one polyolefin that contains reactive groups or polyolefin-containing ionomers, preferably at least one olefin co-, ter- and/or graft polymer that contains reactive groups and/or an ionomer that contains or is based on a polyolefin homo-, co-, ter, and/or graft polymer, wherein the e-modulus of the mixture of (x) and (y) or the e-modulus of the plastic film or plastic film sheet thermoformed from it or the layer (in multilayer plastic films) lies between 50 to 600 N/mm<sup>2</sup>, preferably 70 and 550 N/mm<sup>2</sup>.

The reactive groups of the reactive-group-containing polyolefin and/or of the additional reactive-group-containing elastic polymer or thermoplastic elastomers are preferably carboxyl, hydroxyl, anhydride, amine, amide, isocyanate, epoxy, and/or nitrile groups. The part by weight of reactive groups is 0.2 to 30 wt%, preferably 0.3 to 25 wt%, with respect to the elastic polymers that contain reactive groups (calculated with respect to 100 parts by weight [of the overall content]) or thermoplastic elastomers or polymer mixtures according to (x), which contain reactive groups and 0.2 to 25 wt%, preferably 0.5 to 20 wt%, with respect to the polyolefin(s) or polyolefin-containing ionomer(s) containing reactive groups according to (y).

The polymer(s) and/or thermoplastic elastomer(s) that contain elastic reactive groups is/are completely or partially bonded to the reactive polyolefin, and the bond occurs either completely, or in part, through covalent bonding and/or ionic bonding.

The elastic polymer and/or thermoplastic elastomer which contains reactive groups, according to one embodiment, consists of mixtures of different reactive and non-reactive polymers; here, however, the total content of reactive groups of the mixtures is 0.2 to 30 wt%, preferably 0.3 to 25 wt% (with respect to 100 wt% of the polymer mixture which contains the reactive groups).

According to a preferred embodiment, the plastic film, particularly during the manufacture of the plastic mixture consisting of (x) and (Z), per 100 parts by weight, contains 0.5 to 30 parts by weight, preferably 5 to 25 parts by weight, of another thermoplast (with the

exception, however, of a free reactive-group-containing polyolefin or polyolefinic ionomer or thermoplastic elastomer or elastic polymer).

According to a preferred embodiment, at least the bottom film additionally contains, per 100 parts by weight of (x) and (y)--or of (x), (y) and (z)--0.01 to 6 parts by weight of at least one processing aid or adjuvant, preferably at least one stabilizing agent, antioxidant, UV absorber, lubricant, and/or antistatic agent in a layer of the plastic film or a plastic film sheet consisting thereof.

The plastic molding material or plastic-containing molding material for the manufacture of the bottom film or film sheet, or at least one layer of the bottom film or film sheet, according to a preferred embodiment contains (x) 5 to 80 parts by weight, preferably 10 to 60 parts by weight, of at least one elastic reactive-group-containing polymer and/or at least one thermoplastic reactive-group-containing elastomer or a reactive-group-containing polymer or polymer mixture with a component of elastic polymers of more than 60 wt%, preferably more than 75 wt% (with respect to 100 parts by weight of the elastomer-containing polymer or polymer mixture), (y) 20 to 95 parts by weight, preferably 40 to 90 parts by weight, of at least one reactive-group-containing polyolefin or polyolefin-containing ionomer, preferably at least one reactive-group-containing olefin co-, ter-, and/or graft polymer and/or an ionomer containing or based on a polyolefin co-, ter-, and/or graft polymer, and/or (x), (y), and (z) 0.5 to 30 parts by weight, preferably 5 to 25 parts by weight, of a thermoplast with the exception, however, of a free, reactive-group-containing polyolefin or polyolefinic ionomer or thermoplastic elastomer or elastic polymer, as a plastic component or product consisting thereof, wherein the elastic polymer, elastomer, and/or elastomer-containing polymer or polymer mixture (x) has a different 2% e-modulus--preferably a 2% e-modulus that which is different, in particular lower, by 10 N/mm<sup>2</sup> or more (measured at 20°C) than the reactive-group-containing polyolefin and/or olefinic ionomer--and, with respect to 100 parts by weight (x) and (y), or (x), (y) and (z), the plastic film, plastic film sheet, or layer of the multilayer plastic film or plastic film sheet additionally contains or consists of 0.1 to 30 parts by weight, preferably 0.1 to 20 parts by weight, of at least one filler selected from a group consisting of metal oxides, metal carbonates, metal dioxides, metal hydrates, metal hydroxides, hydrotalcites and/or silicates, and/or an organic filler selected from a group consisting of the fine celluloses, starch and/or fine carbon [carbon-containing substances], calcium carbonate, zinc oxide, magnesium oxide, calcium oxide and/or silicon dioxide, talc, mica or wollastonite, 0.01 to 5 parts by weight, preferably 0.1 to 3 parts by weight, of at least one stabilizing agent, antioxidant and/or UV absorber, 0.01 to 5 parts by weight, preferably 0.01 to 3 parts by weight, of at least one coloring agent and/or color pigment, 0 to 3 parts by weight, preferably 0.1 to 2 parts by weight, of at least one lubricant and/or antistatic agent, and 0 to 20 parts by weight, preferably 1 to 12 parts by weight, of at least one flameproofing agent.

The reactive-group-containing polyolefin (y) consists, according to a preferred embodiment, of a mixture or alloy and/or a co- or graft polymer consisting of an olefin, preferably ethylene and/or propylene, and a carboxylic acid containing at least one unsaturated bond and/or its anhydride, preferably acrylic acid, methacrylic acid, maleic acid and/or maleic acid anhydride, and/or an olefin or polyolefin, preferably ethylene or polyethylene and a compound containing at least one epoxy group, preferably a methacrylate glycidide or a copolymer of one or more of these compounds, or a mixture containing such.

According to another preferred embodiment, the elastic polymer(s) that possess(es) the reactive groups or additional reactive-group-containing compounds consist/consists of a reactive-group-containing ethylene propylene copolymer (EPC), a reactive-group-containing or reactively modified ethylene-propylene diene copolymer (EPDM), reactive-group-containing di-block polymers of styrene with butadiene, with isoprene, with ethylene-butylene, or with ethylene-propylene, preferably of reactive-group-containing styrene-3-block polymers with ethylene butylene and ethylene propylene, an elastic polyester and/or polyethylene ester, polyester amide, polyether amide or copolyester or polyurethane or polyether urethane segment[-containing] polymers.

According to another embodiment, in addition to components (a) and (b) of the bottom film, another peroxide crosslinking agent or a reactive-group-containing organic or polymeric crosslinking agent in parts by weight of 0.1 to 20 parts by weight, preferably 0.3 to 12 parts by weight, are used as well.

According to a preferred embodiment, the reactive-group-containing polymeric crosslinking agent of the bottom film or films, besides *[missing words in original document]* has acrylate and/or methacrylate groups or acrylic acid and/or methacrylic acid groups, and more than 51 wt% ethylene and/or olefin or (CH<sub>2</sub>)- or CH groups, epoxide groups, or epoxy-like groups, preferably glycidyl groups.

The reactive-group-containing polymers and/or ionomers of the bottom film, according to one embodiment, preferably contain alkali metal and/or alkaline earth ions and/or zinc ions.

According to one embodiment, the polymers and/or ionomers that contain reactive groups contain an alkyl acrylate and/or alkyl methyl acrylate with 1 to 8 C atoms.

Component "c" can represent, among other things, a vinyl acetate homo-, co-, or terpolymer or another acid and/or ester-group-containing polymer (EA, EAA, EMA).

The present invention relates, furthermore, to the manufacture of deep-drawable multilayer films, involving a process in which two or more extruders are used for the manufacture of the plastic films, with the extruders being equipped with an extruder sheet die.

According to the invention, in at least one extruder, components a) to c) of the top film and, in at least one other extruder, the components of the bottom film or films are extruded at

temperatures of 150 to 280°C, preferably 180 to 250°C. This involves a process in which a partial reaction or coupling reaction occurs with two or more components of the bottom film during the use of a twin-screw extruder at the extrusion temperature, with a multilayer film being produced as a coextrusion [coextruded] film.

According to another preferred embodiment of the method according to the invention, the Shore D hardness of the top film is set by means of, or with the aid of, at least one plastic that contains a polyolefin or polyolefin groups and has a lower Shore D hardness (measured according to DIN 53505) as well as, optionally, by means of at least one polyolefin-containing or polyolefin-group-containing plastic with a higher Shore D hardness--to a Shore D value that is lower than the Shore D value of the bottom film arranged underneath it by more than 3 Shore D units, preferably by 5 to 10 Shore D units.

The hardness of the top film is set--by selecting the polyolefin-containing and/or olefin group-containing plastics or components with a lower or higher Shore D hardness according to their parts by weight--to a hardness of less than 37 D Shore, preferably 28 to 35 D Shore (measured according to DIN 53505), and the hardness of the bottom film(s) is set to at least 38 D Shore, preferably 39 to 46 D Shore.

According to a preferred embodiment, the top film (on the surface that is opposite to the bottom film) and/or the bottom film (on the surface that is opposite to the top film) are provided with at least one polyacrylate, polymethacrylate, and/or polyurethane-containing diluting agent, preferably one with an organic-chemical-solvent-containing coating using a coating application method, preferably using a spritz, spray, brush, roller, or print drum application method, during which the coating of the top film contains 0.01 to 18 wt/%, preferably 0.1 to 12 wt%, of at least one fine matting agent.

Preferably, at least one antistatic agent and/or at least one adhesion-promoting chemical agent, preferably ultrafine carbon or soot with an average particle size of under 100 nm, preferably between 10 and 80 nm, and/or at least one ultrafine silicic acid or an ultrafine silicon dioxide and/or silicate with an average grain diameter of under 4 µm, preferably under 3 µm, is used for the layer or coating arranged underneath the bottom film.

Furthermore, the present invention relates to the use of the multilayer film alone or in combination with a foam layer, substrate layer, supporting layer, spacer layer, or a lattice or tissue layer for the manufacture of automobile interior trim, preferably for control panels or trim boards for automobiles, side wall parts for automobiles, door trim for automobiles, interior ceiling trim (inside roof lining), interior rear-wall parts for automobiles (rear shelf), and other interior trim parts or objects for automobiles.

Top film examples	1	2
PP block polymer <sup>1</sup>	80	-
PP block polymer <sup>2</sup>	-	60
PP-EPDM <sup>3</sup>	10	10
PP-EPDM <sup>4</sup>	-	10
Ionomer <sup>5</sup>	10	-
Ionomer <sup>6</sup>	-	10

(1) MFI = 0.8 g/10 min 230 °C/21.6 N

(2) MFI = 3.0 g/10 min 230 °C/21.6 N

(3) MFI = 5.0 g/10 min 230 °C/21.6 N

(4) MFI = 30.0 g/10 min 230 °C/21.6 N

(5) Cationic-type zinc MFI 1.0 g/min 190 °C/21.6 N

(6) Cationic-type zinc MFI 3.0 g/min 190 °C/21.6 N

Bottom film examples	1	2	3	4	5
PP block polymer <sup>1</sup>	82	-	80	-	-
PP copolymer <sup>3</sup>	-	20	-	-	-
PP homopolymer <sup>8</sup>	-	-	-	30	20
SEBS <sup>2</sup>	18	-	10	-	-
EPDM-1 <sup>4</sup>	-	50	-	-	-
LLDPE <sup>5</sup>	-	30	10	-	-
EVA <sup>6</sup>	-	-	-	70	-
DHBP <sup>7</sup>	-	-	-	0.2	-
EPDM-2 <sup>9</sup>	-	-	-	-	45
Ionomer <sup>10</sup>	-	-	-	-	35
Tensile stress (at 170 °C) (Pa)	69.10 <sup>3</sup>	40.10 <sup>3</sup>	47.10 <sup>3</sup>	80.10 <sup>3</sup>	130.10 <sup>3</sup>

(1) MFI = 0.8 g/10 min 230 °C/21.6 N

(2) MFI = 3.0 g/10 min 200 °C/50.0 N styrene-ethylene-butylene-styrene

(3) MFI = 6.0 g/10 min 230 °C/21.6 N

(4) MFI = 2.0 g/10 min 230 °C/21.6 N

(5) MFI = 2.0 g/10 min 230 °C/21.6 N

(6) Ethylene vinyl acetate with 28% VAC content

(7) Dimethyl butyl peroxy hexane

- (8) MFI = 4.0 g/10 min 230 °C/21.6 N
- (9) Mooney viscosity 46 ML (2+10) 120 °C
- (10) MFI = 1.0 g/10 min 190 °C/21.6 N ethylene-acrylic acid copolymer

#### Description of symbols

In the enclosed drawings (Figures 1 and 2), several sample embodiments of the multilayer film according to the invention are depicted schematically, wherein the thickness dimensions do not correspond to the actual thicknesses of the layers (this is particularly the case for the thickness of the surface protection layer or paint layer, which in practice lies between 8 and 60  $\mu\text{m}$ , preferably 10 to 50  $\mu\text{m}$ ).

Under (1), the paint or surface protection layer is illustrated, which is preferably applied in the form of a binding agent or solvent such as a synthetic resin to form the surface protection layer.

The bottom film(s) (3) is/are arranged under the top film (2). In addition, in Figure 2, another layer (4) is depicted schematically, with this being a foam and/or substrate layer.

#### Patent claims

1. Deep-drawable multilayer film that consists of a polyolefin-containing top film and at least one polyolefin-containing bottom film that deviates qualitatively and/or quantitatively from the composition of the top film, as well as, optionally, a surface protection layer, characterized in that the top film contains (with respect to 100 parts by weight of plastic of the top film)
  - a) 20 to 80 parts by weight  
of at least one crosslinked PP-EPDM (alloy or mixture of propylene homo- and/or copolymer and crosslinked or partially crosslinked ethylene-propylene diene copolymer)
  - b) 20 to 80 parts by weight  
of an uncured propylene block copolymer or heterophase propylene block polymer with an ethylene-propylene elastomer component in the block or in the chain present at 35 to 75 wt% (with respect to 100 parts by weight of the uncured heterophase propylene block copolymer or propylene block polymer), wherein b is [can be] substituted at 1 to 15 wt%, preferably 2 to 10 wt%, of the same parts by weight of a reactive-group-containing polymer and/or ionomer c, wherein

- c) at least one reactive-group-containing polymer and/or ionomer based on ethylene-methacrylic acid and/or acrylic acid, wherein the acid groups partially contain metal ions or are neutralized by metal ions, and the bottom film or films contain(s) at least two different polyolefins or polyolefin-containing polymers, co-, ter- or block polymers.
- 2. Deep-drawable multilayer film according to Claim 1, characterized in that the polypropylene block of the uncured heterophase polypropylene block polymer consists of a polypropylene homo-, co- and/or block polymer and the elastomer component of the heterophase polypropylene block copolymer is present at 40 to 70 wt% (with respect to 100 parts by weight of the uncured heterophase polypropylene block copolymer).
- 3. Deep-drawable multilayer film according to Claims 1 and 2, characterized in that the bottom film contains at least one propylene homo-, co-, ter-, or block polymer with or without reactive groups and an ethylene-propylene polymer (EPM) and/or ethylene-propylene diene copolymer (EPDM) and/or ethylene-propylene diene copolymer (EPDM) and/or one of components a, b, and/or c of the top film.
- 4. Deep-drawable multilayer film according to one or more of Claims 1 to 3, characterized in that the heterophase propylene block polymer possesses an Mfi 230/2.16 g/10 min of 0.3 to 8, preferably 0.5 to 7.
- 5. Deep-drawable multilayer film according to one or more of Claims 1 to 4, characterized in that dynamically crosslinked or partially crosslinked PP-EPDM, preferably those having a density between 0.83 and 0.96 g/cm<sup>3</sup>, preferably between 0.87 and 0.91 g/cm<sup>3</sup>, are preferably used for the top and/or bottom films as the PP-EPDM.
- 6. Deep-drawable multilayer film according to one or more of Claims 1 to 5, characterized in that the PP-EPDM for the bottom and/or top film has a flexural modulus (initial flexural modulus)--measured according to ASTM D 790 with a 2-mm-thick film sheet--of 80 to 400 MPa, preferably 100 to 300 MPa.
- 7. Deep-drawable multilayer film according to one or more of Claims 1 to 6, characterized in that the bottom film contains at least one partially crosslinked EPDM and a propylene homo-, copolymer or graft polymer with or without reactive groups, a PP-EPDM or dynamically crosslinked or partially crosslinked PP-EPDM with at least one of the components b or c of the top film and/or at least one reactive-group-containing polymer and/or ionomer.
- 8. Deep-drawable multilayer film according to one or more of Claims 1 to 7, characterized in that the bottom film consists of 65 to 90 wt% (with respect to 100 wt% of the plastic component of the bottom film) propylene homo-, co-, ter-, and/or block polymer and 10 to 35 wt% ethylene-vinyl acetate copolymer with a vinyl acetate content of 10 to 40 wt%,

preferably 20 to 35 wt%, wherein a plastic component or a mixture of the plastic alloy is treated with 0.1 to 5.0 wt%, preferably 0.2 to 1.5 wt%, of at least one peroxide or a peroxide group-containing compound.

9. Deep-drawable multilayer film according to one or more of Claims 1 to 8, characterized in that the bottom film consists of 70 to 85 wt%, preferably 75 to 83 wt% (with respect to 100 wt% of the plastic component of the bottom film) propylene homo-, co-, ter- and/or block polymer and 15 to 30 wt%, preferably 17 to 25 wt% ethylene-vinyl acetate copolymer, which contain at least one peroxide or a peroxide-group-containing compound, or are treated therewith.
10. Deep-drawable multilayer film according to one or more of Claims 1 to 9, characterized in that the bottom film contains at least one partially crosslinked EPDM, a propylene homo-, copolymer or graft polymer with or without reactive groups, as well as a block, di-block or tri-block and/or graft polymer containing an additional olefin or polyolefin and/or styrene or polystyrene or that is completely or partially substituted by a modification agent based on a polypropylene-maleic acid anhydride mixture or graft polymer, a polypropylene-acrylic acid mixture or graft polymer, an ethylene-vinyl acetate copolymer, an ethylene-acrylic acid or methacrylic acid copolymer, an ethyleneacrylic acid copolymer, an ethylene-acrylic acid ester copolymer, a glycidyl methacrylate co- or terpolymer, preferably an olefinic or olefin-group-containing glycidyl methacrylate co- or terpolymer and/or and ethylene-glycidyl methacrylate terpolymer or copolymer, an olefinic ionomer, a polyurethane, a polycaprolactone, a polyester ether and/or an polyether amide.
11. Deep-drawable multilayer film according to one or more of Claims 1 to 10, characterized in that the components of the bottom film are selected in such a manner that the bottom film made thereof (with respect to the initial cross-section of the bottom film) has a maximum tensile stress in the range of  $40 \times 10^3$  to  $150 \times 10^3$  Pa (these values are determined at a constant deformation rate at 170°C) and/or that the strain-stress curve runs approximately parallel to the elongation axis at an elongation of over 150%, preferably over 300% elongation, after reaching the maximum [stress-strain].
12. Deep-drawable multilayer film according to one or more of Claims 1 to 11, characterized in that the top film has an average thickness of  
60 to 500  $\mu\text{m}$ , preferably  
100 to 350  $\mu\text{m}$ ,  
and the bottom film or films have an average thickness of  
100 to 2500  $\mu\text{m}$ , preferably



- 500 to 1500  $\mu\text{m}$ , with the bottom film or films having a total thickness that is more than 1.5 times, preferably 2 or more times greater, than that of the top film.
13. Deep-drawable multilayer film according to one or more of Claims 1 to 13, characterized in that the bottom film or films b contains/contain 1 to 60 wt%, preferably 5 to 55 wt%, of at least one partially crosslinked EPDM
    - e) 12 to 30 wt%, preferably 13 to 25 wt%
 

of at least one propylene homo-, copolymer or graft polymer with or without reactive groups, and that, as a plastic component, two or more olefin-containing polymers are contained as residual components, at least one of which represents a reactive-group-containing polymer and/or ionomer.
  14. Deep-drawable multilayer film according to one or more of Claims 1 to 13, characterized in that the propylene or polypropylene content (content of propylene homo-, co-, ter- or block polymer) of the crosslinked PP-EPDM (component a) of the top film, with respect to 100 parts by weight of the PP-EPDM contained in the top film, lies between 60 to 90 wt%, preferably 65 to 85 wt%, and that the EPDM has been crosslinked with a peroxide or a peroxide-group-containing compound and is present at a part by weight (with respect to 100 parts by weight of the PP-EPDM contained in the top film) level of 10 to 40 wt%, preferably 15 to 35 wt%.
  15. Deep-drawable multilayer film according to one or more of Claims 1 to 14, characterized in that the top film has (or is set to) a Shore D hardness that is lower than the Shore D hardness of the bottom film(s) by more than 3 Shore D units, preferably 5 to 10 Shore D units, and/or that the surface tension of the top film is set to a value greater than 40 mN/m.
  16. Deep-drawable multilayer film according to one or more of Claims 1 to 15, characterized in that the top film using at least one plastic with a lower Shore D hardness and at least one component or plastic with a higher Shore D hardness is set to a Shore D hardness of less than 37, preferably to a Shore D hardness of 28 to 35, and the Shore D hardness of the bottom film(s) is at least 38 Shore D, preferably 39 to 48 Shore D.
  17. Deep-drawable multilayer film according to one or more of Claims 1 to 16, characterized in that at least one polyacrylate, polymethacrylate, and/or polyurethane-containing layer is arranged above the top film or on the film surface and/or underneath the bottom film, whose layer thickness is less than the layer thickness of the top film or bottom film, wherein the layer thickness of the coating is less than 50% of the layer thickness of the top or bottom film(s).
  18. Deep-drawable multilayer film according to one or more of Claims 1 to 17, characterized in that the top film and, optionally, partial areas of the bottom film(s) are embossed or

scarred and provided with at least one polyacrylate, polymethacrylate, and/or polyurethane-containing layer or coating that contains 0.01 to 18 wt%, preferably 0.1 to 12 wt%, of at least one fine matting material, and is coated by the application of diluting agent.

19. Deep-drawable multilayer film according to one or more of Claims 1 to 18, characterized in that the polyacrylate, polymethacrylate, and/or polyurethane-containing layer(s) or coating(s) or layer(s) or coating(s) consisting of them (with respect to the plastic component) preferably contain at least one antistatic agent and/or at least one adhesion-promoting chemical agent, preferably ultrafine carbon or soot with an average particle size of under 100 nm, preferably between 10 and 80 nm, and/or at least one ultrafine silicic acid or an ultrafine silicon dioxide and/or ultrafine silicate with an average grain diameter of under 4  $\mu\text{m}$ , preferably under 3  $\mu\text{m}$ .
20. Method for the manufacture of deep-drawable multilayer film according to one or more of Claims 1 to 19, wherein two or more extruders are used for the manufacture of the plastic film, with the extruders being equipped with an extruder sheet die, characterized in that, in at least one extruder, components a to c of the top film, and in at least one extruder the components of the bottom film or films are extruded at temperatures of 150 to 280°C, preferably 180 to 250°C, which involves a process in which a partial reaction or coupling reaction occurs with two or more components of the bottom film, preferably the reactive components with reactive groups or reactive compounds, during the use of a twin-screw extruder at the extrusion temperature, and with the multilayer film being produced as a coextrusion [coextruded] film.
21. Method according to Claim 20, characterized in that the Shore D hardness of the top film is set--by means of or with the aid of at least one plastic that contains a polyolefin or polyolefin groups and has a lower Shore D hardness (measured according to DIN 53505) as well as, optionally, by means of at least one polyolefin-containing or polyolefin-group-containing plastic with a higher Shore D hardness--to a Shore D value that is lower than the Shore D value of the bottom film arranged underneath it, by more than 3 Shore D units, preferably by 5 to 10 Shore D units.
22. Method according to Claims 20 and 21, characterized in that the hardness of the top film is set--by selecting the polyolefin-containing and/or olefin-group-containing plastics or components with a lower or higher Shore D hardness and their parts by weight--to a hardness of less than 37 D Shore, preferably 28 to 35 D Shore (measured according to DIN 53505), and the hardness of the bottom film(s) is set to at least 38 D Shore, preferably 39 to 46 D Shore.

23. Method according to one or more of Claims 20 to 22, characterized in that the top film (on the surface that is opposite the bottom film) and/or the bottom film (on the surface that is opposite the top film) is/are provided with at least one polyacrylate, polymethacrylate, and/or polyurethane-containing and diluting agent, preferably an organic-chemical-solvent-containing coating, using a coating application method, preferably using a spritz, spray, brush, roller, or print drum application method, during which process the coating of the top film contains 0.01 to 18 wt/%, preferably 0.1 to 12 wt%, of at least one fine matting agent.
24. Method according to one or more of Claims 20 to 23, characterized in that at least one antistatic agent and/or at least one adhesion-promoting chemical agent, preferably ultrafine carbon or soot with an average particle size of under 100 nm, preferably between 10 and 80 nm, and/or at least one ultrafine silicic acid or an ultrafine silicon dioxide and/or silicate with an average grain diameter of under 4  $\mu\text{m}$ , preferably under 3  $\mu\text{m}$ , is used for the layer or coating arranged underneath the bottom film.
25. Use of the multilayer film according to one or more of Claims 1 to 19 alone or in combination with a foam layer, substrate layer or a substrate [sic], supporting layer, spacer layer, or a lattice or tissue layer for the manufacture of automobile interior trim, preferably for control panels or trim boards for automobiles, side wall parts for automobiles, door trim for automobiles, interior ceiling trim (inside roof lining), interior rear-wall parts for automobiles (rear shelf), and other interior trim parts or objects for automobiles.

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With 2 page(s) of drawings

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FIGURE 1

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	2
	3

FIGURE 2

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	2
	3
	4

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